Resonant Rayleigh Scattering from an Inhomogeneously Broadened Transition: A New Probe of the Homogeneous Linewidth

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Strong elastic scattering of laser light, up to 200 times background, is found at the exciton resonance of GaAs/AlGaAs multi-quantum-well structures. It is shown to be a consequence of spatial fluctuations of the refractive index due to inhomogeneous broadening of the resonance. The frequency and temperature dependence of the homogeneous linewidth can be obtained from the data.

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Elastic ("Rayleigh") scattering of light in solids usually arises from imperfections in the bulk or on the surface. In measurements of Raman or Brillouin scattering this unshifted signal is generally much larger than the wanted, frequency-shifted, signal. However, crystals can be grown in which this defect scattering is extremely weak. In such crystals we find a strong resonant contribution to the Rayleigh scattering in the vicinity of an inhomogeneously broadened optical resonance. This scattering arises from spatial fluctuations in the local resonant frequency which, because of the strong dispersion near resonance, lead to corresponding fluctuations in the refractive index. In such a case Rayleigh scattering ceases to be a trivial, if annoying, "dirt" effect, and can give useful information on the nature of the resonance, in particular on its homogeneous linewidth. This scattering is distinguished from resonant fluorescence by the fact that its spectral and temporal dependence is strictly that of the exciting laser. To our knowledge such elastic scattering from an inhomogeneously broadened transition has not been reported before. It is quite distinct from the resonantly enhanced Rayleigh scattering by density fluctuations seen, for example, in Hg vapor.[1]

We have observed this scattering in the vicinity of the lowest, "heavy," exciton resonance of GaAs/AlGaAs multi-quantum-well (MQW) structures, grown by molecular-beam epitaxy. These structures consist of about 100 alternating layers of GaAs and $Al_{0.3}Ga_{0.7}As$. The AlGaAs layers are 200 Å thick; the GaAs layer thickness L_z is chosen to be between 50 and 200 Å. In these structures the exciton is two dimensional and its energy depends on L_z. Its linewidth (full width at half maximum) Γ_x is predominantly inhomogeneous at low temperatures and arises from spatial fluctuations in L_z.[2] In a previous publication[3] it was shown, by hole-burning experiments, that on the low-energy side of the line the homogeneous linewidth Γ_h is much less than Γ_x.

Because etching to remove the substrate introduces excessive surface scattering, all our measurements were made on as-grown samples. The sample was carefully cleaned and the laser focused to a spot well away from the edges. The light scattered from this spot was focused on a pinhole in front of the slit of a high-resolution spectrometer. Figure 1 shows a series of cw "emission" spectra (fluorescence plus scattered light) obtained by scanning the spectrometer through the exciting dye-laser frequency ω, for 23 different values of ω, indicated by the arrows. The laser power was held constant at about 10 mW/cm^2 and the spectral resolution here was 0.1 meV. When the laser frequency is set off resonance there is a weak sample-dependent Rayleigh line due to defect scattering. When it is set close to resonance the Rayleigh line is dramatically enhanced, reaching a peak some 200 times the off-resonance defect scattering of a good quality sam-

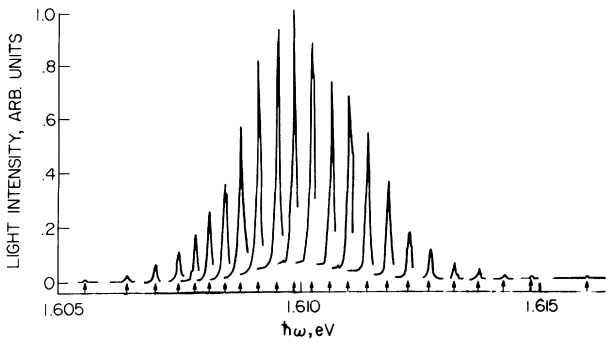

FIG. 1. Scattered light spectra (cw) for an MQW structure with $L_z = 51$ Å, obtained at the 23 different laser frequencies ω indicated by the arrows. $T = 5.7$ K. The broad background is exciton fluorescence. The laser intensity is the same for each spectrum.

ple. On the low-energy side of the resonant line in each spectrum there is a shoulder due to nonresonant exciton luminescence. Similar results are obtained for a sample with $L_z = 208$ Å.

Within our highest resolution (0.05 meV) the scattered light has the same spectral profile as the laser. It is at least 80% polarized and is isotropic. We also made measurements with the dye laser mode locked, giving 10-psec pulses. Our detection system had sufficient time resolution to detect a decay time down to 30 psec. The resonantly scattered signal was found to have a time profile indistinguishable from that of the laser pulse apart from a weak, relatively long-lived component (~ 200 ps for $L_z = 208$ Å) due to exciton fluorescence, which represents less than 10% of the integrated signal. These results show that the resonant signal is due not to fluorescence, but to a true scattering process. While there is a change of reflectivity at the GaAs:AlGaAs interfaces near resonance, this is only of order 0.1%, too small to account for the observed effect.

We attribute the scattering to spatial fluctuations of the dielectric response. The inhomogeneously broadened line is the envelope of homogeneously broadened packets of excitons, each packet corresponding to a particular local value of the layer width L_z. When the laser is in resonance with one particular packet, it is off resonance elsewhere. Because of the strong resonant dispersion this produces corresponding spatial fluctuations in the dielectric response at the laser frequency. Their strength increases inversely as the homogeneous linewidth Γ_h.

We assume the local resonant frequency $\Omega(\vec{r})$ to be a random variable with distribution $g(\Omega)$, whose width is γ. The results are insensitive to the form of $g(\Omega)$ so long as $\gamma \gg \Gamma_h$, but for definiteness we take $g(\Omega)$ to be Lorentzian, so that $\gamma = \Gamma_x - \Gamma_h$, where Γ_x is the observed exciton linewidth. The dielectric response function at any point is given by[4]

$$\epsilon(\omega,\vec{r}) = \epsilon_\infty[1+\Lambda^2/(\Omega^2-\omega^2-i\omega\Gamma_h)], \quad (1a)$$

where ϵ_∞ and Λ^2 are constants defined by Loudon.[4] When $|\Omega-\omega| \ll \Omega$ this simplifies to

$$\epsilon(\omega,\vec{r}) = \epsilon_\infty + \epsilon_\infty\omega_{LT}/\{[\Omega(\vec{r})-\omega]+\tfrac{1}{2}i\Gamma_h(\Omega)\}. \quad (1b)$$

Here $\omega_{LT} = \Lambda^2/2\Omega$ is the longitudinal-transverse splitting of the exciton, and Γ_h is assumed to be a function only of Ω. If the correlation length ξ is much less than the wavelength λ, the scattered intensity due to spatial fluctuations in ϵ is isotropic, as observed. It is proportional to $S(\omega)$ where[1,5]

$$S(\omega) = \langle|\epsilon(\omega)-\langle\epsilon(\omega)\rangle|^2\rangle = \langle|\epsilon(\omega)|^2\rangle - |\langle\epsilon(\omega)\rangle|^2, \quad (2)$$

where $\langle f(\omega)\rangle \equiv \int g(\Omega)f(\Omega-\omega)d\Omega$ for any f. Substituting from Eq. (1b), and using $\alpha(\omega) = 2\pi\lambda^{-1}\epsilon_\infty^{-1/2} \times \langle \mathrm{Im}(\epsilon)\rangle$ for the absorption coefficient, we find, after some algebra,

$$S(\omega) = \pi^{-1}\lambda\epsilon_\infty^{3/2}\omega_{LT}\alpha(\omega)[\Gamma_h(\omega)^{-1} - \Gamma_x^{-1}], \quad (3)$$

where we have assumed that $d\Gamma_h/d\omega \ll 1$. Integrating over the total GaAs thickness d, with allowance for reabsorption, we obtain for the backscattered intensity

$$I_s(\omega) = K(1-e^{-2\alpha(\omega)d})[\Gamma_h(\omega)^{-1} - \Gamma_x^{-1}], \quad (4)$$

where K is an as yet undetermined constant.

The above analysis breaks down if $\Gamma_h < 2\omega_{LT}$, since the response near resonance becomes metallic [i.e., $\mathrm{Re}(\epsilon) < 0$]. The scattering then depends on the shape of the scattering elements, and a full analysis becomes extremely difficult. We can, however, estimate the limiting scattering, as $\Gamma_h \to 0$, as follows. We treat the scattering elements as perfectly conducting disks of radius ξ and volume $V = \pi\xi^2 L_z$, embedded in a dielectric with $\epsilon = \epsilon_\infty$. Their dipole moment per unit field is[6] $P = 16\epsilon_\infty\xi^3/3$. The absolute value of $\epsilon - \epsilon_\infty$ which would produce the same dipole moment is $|\epsilon - \epsilon_\infty|_{max} = P/V = 16\epsilon_\infty\xi/3\pi L_z$. Thus $I_s(\omega)$ does not diverge as $\Gamma_h \to 0$, but reaches a maximum value corresponding to an effective value of $\Gamma_h = \Gamma_0$, where

$$\Gamma_0 = 2\epsilon_\infty\omega_{LT}/|\epsilon-\epsilon_\infty|_{max} \sim L_z\omega_{LT}/\xi. \quad (5)$$

The corresponding relation for the three-dimensional case is $\Gamma_0 \sim \omega_{LT}$. The extra factor L_z/ξ accounts for the enhanced resonant scattering of these MQW structures, over, say, that of bulk GaAs.

Figure 2(a) shows the frequency dependence at 5.6 K of the absorption coefficient,[7] the nonresonant luminescence, and the scattered intensity, for a sample with $L_z = 51$ Å (nominal). Figure 2(b) shows the frequency dependence of $\Gamma_h(\omega)$ at two temperatures, obtained by substituting the measured $\alpha(\omega)$ and $I_s(\omega)$ in Eq. (4). Γ_0 is taken to be the limiting value of Γ_h at low ω and T.

The absolute value of Γ_h [or, equivalently, the value of K in Eq. (4)] can be estimated from Eq. (5). Loudon[5] gives $\omega_{LT} = (\lambda/2\pi^2\epsilon_\infty^{1/2})\int\alpha(\omega)d\omega$, from which $\omega_{LT} = 3\times10^{11}$ s^{-1}; while Petroff *et al.*[8] have observed steps in the GaAs/AlGaAs interface whose spacing suggests that $\xi \sim 200$ Å. Hence, from Eq. (5), $\Gamma_0 \sim 7\times10^{10}$ s^{-1} for $L_z = 51$ Å. This

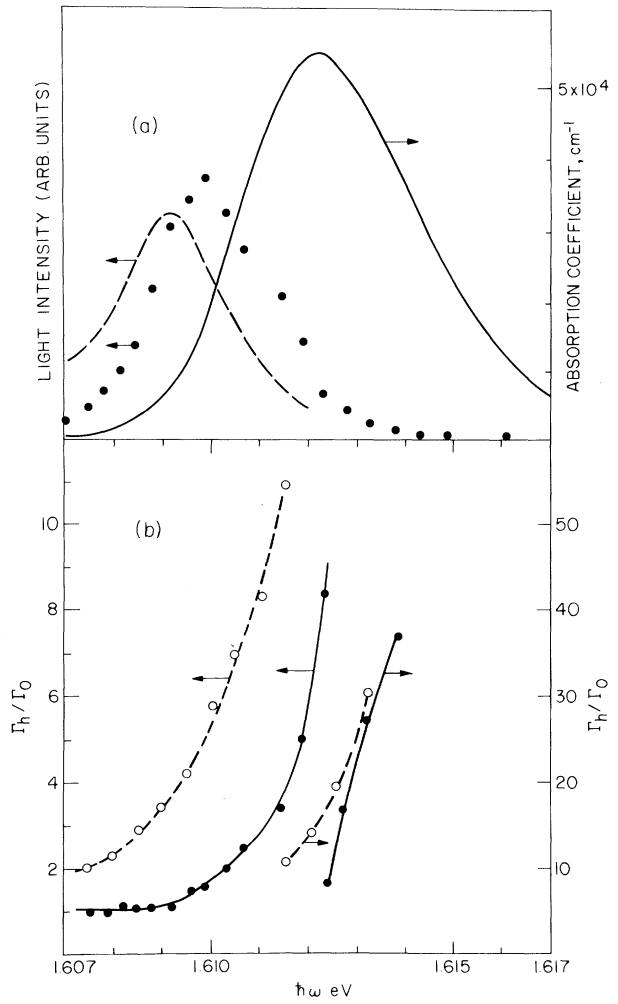

FIG. 2. (a) Full line: absorption spectrum of the sample shown in Fig. 1. The peak corresponds to the 1S "heavy exciton resonance, of width Γ_x. Dashed line: nonresonant fluorescence spectrum. Points: peak scattered intensity at 5.7 K. (b) $\Gamma_h(\omega)/\Gamma_0$ deduced from the data of (a) via Eq. (4). $\Gamma_0 \sim 7\times 10^{10}$ s^{-1} is the limiting value of Γ_h at low ω, and T. Closed circles: T = 5.7 K; open circles: T = 14 K.

is in reasonable agreement with the hole-burning results[3,9] on the same system.

If the laser intensity is increased, I_s increases proportionally until a power density of about 10 W cm^{-2} is reached, where I_s levels off. This drop in scattering efficiency implies an increase in Γ_h, presumably due to exciton-exciton interaction. The exciton density at 10 W cm^{-2} is 10^9 cm^{-2} per layer. A similar increase in Γ_h has been seen in hole-burning experiments.[3]

While an accurate theoretical treatment of the region where $\Gamma_h < \omega_{LT}$ would have to be much more sophisticated, our crude analysis is sufficient to show that in GaAs MQW structures Γ_h increases sharply with exciton energy and temperature. A fuller account of the energy and temperature dependences of Γ_h, which show some evidence for exciton localization, will be published elsewhere.

We conclude that resonant Rayleigh scattering can give useful information on the homogeneous contribution to an inhomogeneous linewidth. The method has the advantage over other techniques, such as hole burning and photon echo, of being usable at very low excitation intensity. On the other hand, a large correlation length ξ is necessary to produce appreciable scattering, since in Eq. (4) $K \propto \xi^3$. Thus, this method is unlikely to be useful in impurity systems such as ruby. It might, however, be applicable to singlet excitons in molecular crystals.

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[7]The absorption spectrum was measured on a sample from which the substrate had been etched away, and was corrected for the strain thereby introduced by comparison of the excitation spectra of the etched and as-grown samples.

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